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Search for Superheavy Elements in Damped Collisions Between ^{238}U and ^{248}Cm

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Abstract:

Upper limits for the production of superheavy elements (SHEs) in damped collisions between ^{238}U projectiles and ^{248}Cm targets were measured. This reaction is believed to permit a closer and more widespread approach to the predicted island of stability near Z=114 and N=184 than any practical fusion reaction. Aqueous and gas phase chemistry techniques were used to isolate SHE fractions. The fractions were counted for spontaneous fission activity, fragment kinetic energies and neutron multiplicities. Cross-section limits for half-lives from hours to several years are $<4x10^{-35}$ cm^2.

The sensitivity of the predicted[1,2] stability against spontaneous fission (SF) of superheavy elements (SHEs) near Z=114 dictates a very small choice of nuclear reactions which can yield a product as close as possible to the 184-neutron shell. Below 184 neutrons, according to Randrup's calculations[1], the fission barrier heights decrease precipitously becoming only 3 to 4 MeV at 178 neutrons. The much favored $^{48}Ca+^{248}Cm$ fusion reaction would produce element 116 with 178 neutrons if only two neutrons are evaporated. The repeated failure of this approach[3,4,5] may be connected[1] with a non-detectable, short half-life and an insufficient survivability of the compound nucleus due to fission.

In order to minimize yield losses by fission during the deexcitation of the compound nucleus, it is imperative to choose a reaction which comes as close as possible to the 184-neutron shell, where the fission barriers are highest. Previous studies of damped, binary collisions with large mass transfer such as $^{238}U+^{238}U$ collisions[6-8] and $^{238}U+^{248}Cm$ collisions[9] and estimates[10] of the most probable N/Z ratios of the resulting fragments at Z = 114 have suggested that their isotopic distribution (FWHM≅3 amu) should be centered at N = 182 with N = 184 fragments being abundantly included in the dispersion. Most of these neutron-rich superheavy fragments will decay by prompt fission, but a small fraction originating with very low excitation energies may survive. The distribution of excitation energies of Z = 114 fragments in $^{238}U+^{248}Cm$ collisions has been estimated[9] on the basis of Q-value measurements in the very similar $^{238}U+^{238}U$ reaction[6]. For excitation energies of 25-35 MeV (which are possible due to the large negative Q_{gg}-value) one obtains[9] a cross section of 10^{-32} cm^2 which is a factor of 30 higher than the same estimate for the $^{238}U+^{238}U$ reaction. Further, careful studies of the fission properties[11] of the heaviest (superheavy) fragments produced in these collisions did not reveal any evidence for non-equilibrium fission. Also, the production of surviving actinides both in ^{238}U on ^{238}U and ^{248}Cm collisions[7,9] was not incompatible with equilibrium statistical decay of the produced heavy fragments. In view of these positive aspects, and in particular, in view of the troublesome outcome of the complete fusion approach with the $^{48}Ca+^{248}Cm$ reaction, it was felt that a major experimental effort with the $^{238}U+^{248}Cm$ reaction was in order.

The experiments were carried out at the UNILAC accelerator. The integral particle numbers per experiment varied from $2x10^{14}$ to $2x10^{16}$. Targets of ^{248}Cm, containing 4.2-5.2 mg/cm^2 ^{248}Cm (97% isotopic purity), were produced by evaporation[12] of the metal onto Mo foils of 4.6 mg/cm^2 areal density which were mounted in a specially

designed target and recoil chamber.[13] Before the beam entered the ^{248}Cm target, Mo windows and N_2-cooling gas reduced the energy to 7.30 MeV/amu. Beam energy measurements were performed by inserting a surface barrier detector directly into a low intensity beam. Extensive studies of target failure mechanisms[12-14], as well as details of the extensive safety measures[15] including on-line control of target temperatures, wobbler amplitudes, cooling gas flow-rates and purity, peak beam intensities, are described elsewhere. The target thicknesses were sufficient to further reduce the beam energy E to the Coulomb barrier B (1.17 E/B 1.01). Reaction products emitted within laboratory angles of 55° were stopped in a Cu foil, or in Kr-gas in case of on-line experiments. The chemical separations were based on four different predictions about the chemical properties of SHEs:

(i) Elements 112 through 116 should be volatile in their elemental state at temperatures up to 1000°C ('Pb-like' and 'Hg-like')[16].

(ii) Some elements such as 112, 114, and 118 may even be gaseous at room temperature ('Rn-like')[17].

(iii) Elements 108 through 116 should form strong anionic bromide complexes in aqueous solution ('Pt-like')[18].

(iv) There is the prediction of a strong affinity of SHEs 108 through 116 to sulfur or sulfur compounds[19].

In order to cover the predictions listed above, the Cu catcher foil was processed after bombardment as follows: First, volatile 'Hg-like' and 'Pb-like' elements were evaporated from the Cu-foil by heating in a closed quartz apparatus in Ar/H_2 stream at about 1000°C and transported through an isothermal chromatography tube kept at 600°C ('Hg-like') and 900°C ('Pb-like'). These fractions were condensed separately on thin, cooled Ni-foils coated with evaporated Pd, as suggested by Eichler[20]. Also, provisions were made to condense 'Rn-like' noble gases in a cryogenic chamber[21]. The residual copper was dissolved with a HBr/Br_2-solution to form bromide complexes ('Pt-like') which were separated on two cation exchange colums from the bulk of other reaction products. The final sample was prepared on a thin carbon substrate by evaporation to dryness. During dissolution of the copper and during the evaporation steps, volatile bromides were condensed and subsequently extracted by diethyl-dithiophosphoric acid (DTP)[19] and mounted on thin carbon substrates by evaporation ('DTP I' and 'DTP II'). Chemical yields were estimated on the basis of tracer experiments using the lighter homologs of SHEs. These were 63% ('Hg-like'), 90% ('Pb-like'), 80% ('Pt-like), 55% ('Rn-like'), and 60% (DTP-fractions). Counting of

the off-line chemical fractions started between one and several hours after end of bombardment in a low background detection system which registered single and coincident fission fragments, their kinetic energies, and the number of neutrons per fission using surface-barrier detector pairs inserted into a ^{3}He neutron counting system[22]. 'Rn-like' samples were counted for single fission fragments only. In an on-line gas-sweeping experiment 'Rn-like' products recoiling from the target were stopped in Kr-gas and transported through a filter system to a cryogenic chamber where they were condensed on a cold surface (-150°C) in front of a single surface barrier detector. The transport and counting efficiency of this system was about 55 percent for gaseous nuclei decaying by spontaneous fission. To estimate the time between formation and observation by the annular detector (transport time), we have taken the volume of the recoil chamber, capillary, fittings, etc. and divided by the flow rate of 155 cm^{3}-min^{-1}. This gave a value of one minute.

None of our manyfold experiments revealed evidence for the production of SHEs above the cross section limits as a function of half-life that are depicted in Fig. 1. Detailed information on our results is contained in Table 1 where counting times and registered events for each fraction are listed. Single fragment events were observed relatively frequently. The rate at which these signals were observed is compatible with the known background level for this type of event in the counting system.

Evidence against the association of these events (with or without neutron coincidences) with fission can be gained from a number of considerations.

i) Their pulse-height distribution is peaked at 35-40 MeV. None of the necessary corrections (pulse height defect, energy loss in the substrate) would increase these energies anywhere near the expected values for true fission fragments.

ii) The distribution of diffusion times of the neutrons in paraffin before they are registered in one of the ^{3}He counters exhibits a difference from that measured for ^{248}Cm fission in that events with long diffusion times (>100μs) are more abundant.

iii) The average rate of these events per surface barrier detector pair was about 0.005 per day when the normal shielding of the counter against cosmic rays was present (Footnote e to Table 1). This rate increased to about 0.03 per day, that is by factors of 2 to 10, during a construction period when 3 m of soil was removed from the underground laboratory (Footnote f to Table 1).

iv) The addition of two large area plastic detectors[22] to the 4π fission counter, which registered muons above and below the 4π counter in coincidence with fission events, indicated that these events are frequently (but not always) associated with a muon penetrating one or both plastic detectors.

Thus, it seems plausible that the majority of the events listed in the colums $E_{1(2)}$ and $E_{1(2)}{}^{+n}$ are associated with the interaction of muons with the surface-barrier detectors. Spontaneous fission as a source of these events can be largely ruled out on the grounds of the observations mentioned above, and, by looking at the total detection efficiency for fission fragments vs. the efficiency to detect both fragments in coincidence which are 60% and 54%, respectively. The complete absence of binary fission fragment coincidences in most chemical fractions is in clear conflict with the singles rates, unless the origin of the latter events has nothing to do with spontaneous fission.

Thus, we are left with at most four coincident events which might indicate true fission occuring in the first DTP fraction and in the main sample from the aqueous chemistry ('Pt-like'). Unfortunately, the latter fractions are by far the most likely ones, out of all the chemical fractions, to contain minute amounts of the spontaneously fissioning ^{248}Cm target material. As far as neutron multiplicities can be deduced from these few events, one would guess from the results in Table 1 that the neutron multiplicity of the spontaneous fission emitter in the 'Pt-like' fraction is not very high, i.e. not incompatible with ^{248}Cm ($\bar{\nu}$=3.15). Decay of this isotope is then seen as a much more reasonable explanation for these events than the presence of long-lived superheavy elements. Nevertheless, we have calculated the limits shown in Fig. 1 for the 'Pt-like' fraction on the basis of two registered events which we take as six events from Poisson statistics at the 95% confidence level.

More interesting, at first sight, is the binary fragment coincidence observed in the first DTP fraction indicating (with pulse height defects and the penetration of the substrate foil by one fragment taken into account) a total kinetic energy of about 290 MeV and, in addition, the unusually high number of 12 registered neutrons. However, interest in this event fades when we consider fission Q-values: For symmetric binary fission, the 290 MeV of kinetic energy leaves only some 35 MeV of excitation energy for the fragments, definitely too little to account for the evaporation of, presumably, far more than 12 neutrons from the fragments. On the other hand, the assumption of three or four equally sized fragments in the fission exit channel would increase the fission Q-values to roughly 400 MeV. If we assume that these three or four fragments were all detected by the pair of surface-barrier detectors the total kinetic energy would be roughly 300 MeV and this now leaves about 100 MeV of fragment excitation energy to be carried away by presumably many more than 12 evaporated neutrons. Seemingly, this is somewhat more reasonable but still improbable. Furthermore, the distribution of diffusion times for the 12 registered neutrons appears to be a typical for true fission because it shows a statistically significant clustering of diffusion

times > 100μs (10 neutrons) while for true fission one observes an exponential decrease of this probability distribution with only a 25% probability for diffusion times > 100μs.

We feel these arguments make the assignment of this particular event to the decay of a superheavy nucleus highly questionable. However, because of the unprecedented nature of the signal, an alternative explanation in terms of electronic noise or cosmic-ray induced reactions in the detectors, is not readily at hand. The limit shown in Fig. 1 for the first DTP fraction represents an upper limit of 6 events which is compatible with two registered eventsat the 95% confidence level.

The reasons for the failure of our approach to synthesise and detect SHEs in damped $^{238}U+^{248}Cm$ collisions may be manyfold: (i) The probability of transferring a large number of nucleons and < 30 MeV of excitation energy (estimated[9] to be of the order of 10^{-32} cm^2) may be too small to offer appreciable yields of surviving SHEs, even though we should be capable, with this approach, of reaching neutron-rich areas in the 'Island of Stability' where the fission barriers are higher and the chances of surviving their formation may be greater than in the complete fusion reactions tried earlier. (ii) Damped collisions may produce the SHE fragments in somewhat deformed shapes even at the lowest excitation energies. If these deformations are larger than the nearly spherical saddle point shapes [1,2] of SHEs, there may not be any driving force towards the spherical ground state and the compound state may be lost to prompt fission. (iii) The spontaneous fission half-lives for surviving SHEs are shorter than the limits established through chemical separations in the present experiments.

While (i) and (ii) are possibilities that cannot be altered by the experimenter, it is possible, but exceedingly difficult and costly, to develop ultrasensitive techniques for the rapid (to a μs), future identification of spontaneous fission nuclides coming from damped transfer reactions. For the time being, we must face the fact that production and identification of SHEs with half-lives from fractions of a day to several years by use of $^{238}U+^{248}Cm$ collisions failed at cross section limits of the order of $10^{-34}cm^2$.

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Table 1: Events from various chemical fractions recorded in the surface barrier detecors. The electronic threshold for fission-fragment pulses was set at 25 MeV. Detection efficiencies were 60 % for fission fragments, 54% for fragment-fragment coincidences, and 26 % for the detection of a single neutron.

Chemical fraction	Net counting time (d)	Observed events [a]				
		t(d)	$E_{1(2)}$	$E_{1(2)}$+n	E_1+E_2	E_1+E_2+n
'Rn-like' [b]	72	0	66	---	---	---
		0	77			
'Hg-like' [c]	936 [e]	168		37(1)		
		181	37			
		660	54			
		695		69(1)		
		815	35			
	119 [f]	233	34			
		235	39			
		242		44(3)		
		272	34			
		347		29(1)		
'Pb-like' [c]	326 [e]	176		33(2)		
	119 [f]	277		31(1)		
		310		53(1)		
		353		31(1)		
DTP I [d]	944 [e]	46	36			
		741		49(2)		
		960	31			
		1041	39			
		1041	29			
		1099			66+101	
	119 [f]	282		33(1)		
		304				150+125(12)
		305		75(1)		
		319	37			
DTP II [d]	326 [e]	146	33			
		231	28			
	119 [f]	301	43			
		309		32(4)		
		311		29(1)		
		317		32(2)		
'Pt-like' [d]	936 [e]	38	34			
		165				70+59(2)
		168		59(1)		
		186	49			
		188			32+32	
		356	54			
		359		35(1)		
		451	28			
		897	30			
		948	89			
	119 [f]	278	34			
		334	98			
		341	47			

Footnotes to Table 1

a Listed are:

t(d)	time (in days) after end of bombardment (July 8, 1981)
$E_{1(2)}$	single events in one fission-fragment detector
$E_{1(2)}+n$	coincidence between a single fission-fragment detector and the neutron counter
E_1+E_2	coincident fission fragments in two opposed fragment detectors
E_1+E_2+n	coincident events in two opposed fission-fragment detectors and the neutron counter

Given for each event are the fission-fragment kinetic energies in MeV and the number of coincident neutrons in parentheses. Underlined energies refer to events registered in the rear detector signifying that the fission fragment was penetrating the substrate foil. The kinetic energies are not corrected for pulse-height defects (5-8MeV) nor for the energy loss in the substrate foil if the fragment was penetrating the foil, e.g. >25-30 MeV for the Ni-substrates, and >4 MeV for the carbon foils. Energy losses in the case of the aqueous chemistry fraction are unknown but estimated to be substantial due to the non-negligible thickness of the sample.

b Condensed on the cold head of a cryopump, counted (outside the neutron multiplicity counter) for single fission-fragment pulses only

c Condensed on rolled Ni foil of 500 $\mu g/cm^2$ thickness coated with 30 $\mu g/cm^2$ of Pd by vacuum evaporation

d On carbon substrate (35 $\mu g/cm^2$) with a 25 $\mu g/cm^2$ evaporated coating of Au

e Measurement in the 4π neutron multiplicity counter under normal background conditions

f Measurement in the 4π neutron multiplicity counter with less cosmic ray shielding than normal

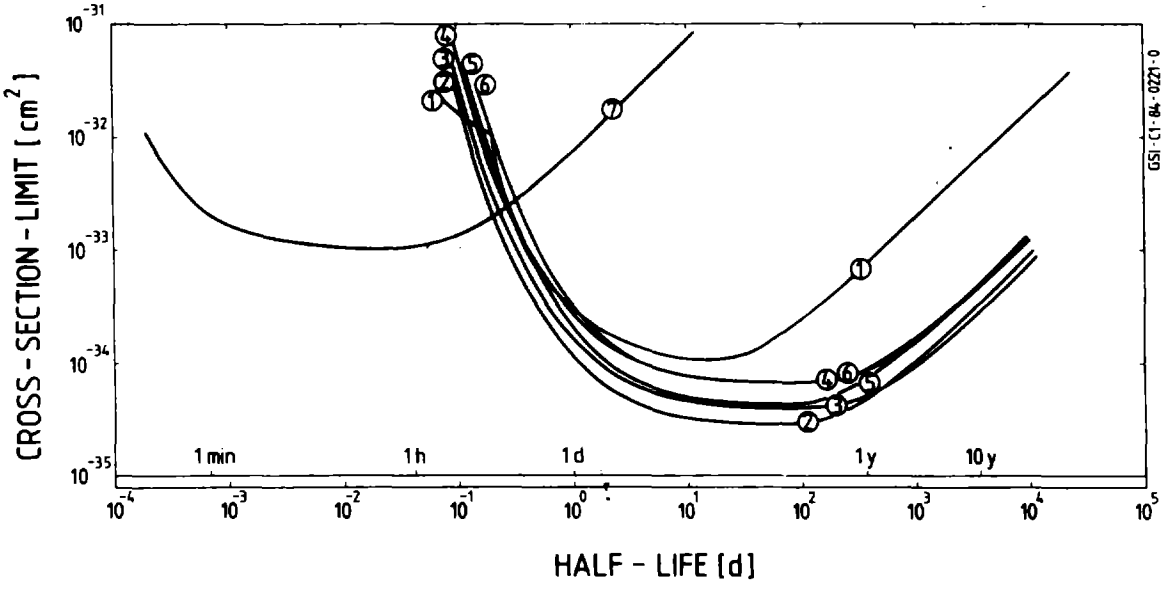

Fig.: 1 Upper limits (at 95% confidence level) for the production cross sections of superheavy nuclei in the reaction of ^{238}U with ^{248}Cm in the energy range 6.2 - 7.3 MeV/amu as a function of the half-life. The curves refer to the following chemical fractions: ① 'Rn-like' (off-line), ② 'Pb-like', ③ 'Hg-like', ④ 'Pt-like', ⑤ DTP II, ⑥ DTP I, and ⑦ 'Rn-like' (on-line).